VOLUME 79 SEPARATE No. 239

PROCEEDINGS

AMERICAN SOCIETY
OF
CIVIL ENGINEERS

AUGUST, 1953

RADIOISOTOPE REMOVAL IN MODERN WASTE TREATMENT

by Warren J. Kaufman and Gerhard Klein

SANITARY ENGINEERING DIVISION

{Discussion open until December 1, 1953}

Copyright 1953 by the AMERICAN SOCIETY OF CIVIL ENGINEERS
Printed in the United States of America

Headquarters of the Society
33 W. 39th St.
New York 18, N. Y.

PRICE $0.50 PER COPY

THIS PAPER

--represents an effort of the Society to deliver technical data direct from the author to the reader with the greatest possible speed. To this end, it has had none of the usual editing required in more formal publication procedures.

Readers are invited to submit discussion applying to current papers. For this paper the final closing dead line appears on the front cover.

Those who are planning papers or discussions for "Proceedings" will expedite Division and Committee action measurably by first studying the printed directions for the preparation of ASCE technical papers. For free copies of these directions—describing style, content, and format—address the Manager, Technical Publications, ASCE.

Reprints from this publication may be made on condition that the full title of paper, name of author, page reference, and date of publication by the Society are given.

The Society is not responsible for any statement made or opinion expressed in its publications.

This paper was published at 1745 S. State Street, Ann Arbor, Mich., by the American Society of Civil Engineers. Editorial and General Offices are at 33 West Thirty-ninth Street, New York 18, N. Y.

RADIOISOTOPE REMOVAL IN MODERN WASTE TREATMENT

Warren J. Kaufman[1], J. M. ASCE, and Gerhard Klein[2]

SYNOPSIS

The development of nuclear power and the utilization of radioisotopes in research and industry are resulting in ever-increasing problems of radioactive waste disposal. Economical processes for the decontamination of these wastes and the concentration of the radioactive components into residues of minimal volume are essential to the future development of the atomic energy industry. This paper reports on a laboratory study of the effectiveness of the activated sludge process in removing the radioisotopes of phosphorus, iodine, and strontium from liquid wastes. The influence of the significant parameters, determining the degree of removal of these isotopes, has been evaluated and the limitations of biological processes for radioactive waste treatment delineated.

INTRODUCTION

In the field of radiological health, the sanitary engineer is faced with challenging and highly complex problems associated with the disposal of liquid radioactive wastes. When classified with respect to origin, these wastes fall into two general categories: those produced by the atomic energy industry, as represented primarily by the United States Atomic Energy Commission, and the liquid wastes of the numerous laboratories and hospitals employing radioisotopes for research and therapeutic purposes. One of the major waste problems of the Atomic Energy Commission arises from the vast quantities of radioactive materials originating as by-products of plutonium manufacture. Such wastes are the inevitable result of nuclear fission and the application of the nuclear reactor as an instrument of power production. These so-called "Fission Products" offer not only numerous attractive possibilities for industrial and scientific application, but also present concomitant problems of preventing all but minimal quantities from reaching man's water and atmospheric environments.

Radioactive wastes have widely differing physical and chemical characteristics which may be determining criteria in selecting a satisfactory treatment process. The treatment of a radioactive laboratory waste by coagulation, for example, has required that the reaction be carried out under highly alkaline conditions in order to overcome the peptizing action of sequestering agents employed as decontaminants. Biological treatment has proven satisfactory for the removal of plutonium from laundry wastes. In other wastes, primarily inorganic in nature, it will be necessary to add supplementary

1. Asst. Prof. of San. Eng., University of California, Berkeley
2. Research Engr., San. Eng. Research Project, University of California, Richmond, California

nutrients if biological systems are to be supported. By definition, the single common feature of radioactive wastes is that they contain elements having unstable nuclei which emit ionizing radiations in a predictable but unchanging fashion. Within the limits of our present knowledge, only the passing of time will bring about the decay of radioisotopes. The rate of this decay may be expressed in terms of the half-life, or time necessary for one half of a quantity of an isotope to undergo radioactive decay.

With the above concepts in mind, it is apparent that only three general methods of handling the radioactive waste problem are available: (a) concentration and controlled confinement, (b) dispersion and dilution to such low concentrations that radiological health hazards are reduced to a minimum, and (c) industrial, medical, or scientific utilization. Major criteria in the concentration of radioisotopes are the volume occupied by the final residue and the cost of extended storage of that residue. Dilution may be accomplished either by discharge into large bodies of water or by the addition of sufficient quantities of stable isotopes to avoid subsequent concentration of the radioactive forms by natural biological or physical processes. Utilization is by far the most attractive method from the standpoint of economy. Unfortunately, utilization in turn may be expected to bring about a multiplicity of problems for regulatory agencies within state and local governments.

Scope of Investigation

The major objective of the study reported herein was to investigate the application of conventional biological treatment processes to the concentration of radioisotopes from liquid wastes. Secondary objectives were to study the mechanisms of biological uptake of ionic materials and to ascertain the role of the municipal biological sewage treatment plant in bringing about possible undesirable concentration of radioisotopes. The activated sludge process was selected for the initial study as one lending itself best to the investigation of the fundamental aspects of biological assimilation. Activated sludge systems may be resonably well identified by physical and chemical determinations, particularly with respect to the concentration of active sludge solids, whereas trickling filter slimes are far less homogeneous and definitive in character.

In becoming cognizant of the great many parameters that might influence the removal and concentration of radioisotopes, it was deemed desirable to make two simplifications in the experimental methodology. Individual radioisotopes fo known chemcial form were selected for the study rather than a mixture of radioactive materials such as the gross fission products. A synthetic sewage, as developed by Weinberger[1], was employed as the basic substrate for the development of the activated sludges. The chemical and sanitary compositions of this "standard substrate" are given in Table 1. The synthetic medium offers the advantage of reproducibility but presents the inherent objection of not being natural, particularly with respect to trace elements and normal sewage biota. This objection was overcome by the periodic addition of small quantities of settled domestic sewage.

The radioisotopes selected for study were phosphorus 32 (p^{32}), strontium 89 (Sr^{89}) and iodine 131(I^{131}). They were obtained from the Oak Ridge National Laboratory as separated isotopes of high radiochemical purity. In all instances, the stable salts present were insufficient to appreciably alter

1. "Nitrogen Metabolism in the Activated Sludge Process," by Leon W. Weinberger, Sc.D. Thesis, Massachusetts Institute of Technology, May, 1949.

Table 1. Composition of Standard Synthetic Substrate

Constituents in Milligrams per Liter of Tap Water		Average Tap Water Analysis ppm	
Nutrient Broth	100	Sodium	4.0
Urea	30	Potassium	0.8
Castile Soap	50	Calcium	4.0
Soluble Starch	100	Magnesium	3.3
Diatomaceous Earth	25	Alkalinity	20.5
NaCl	30	Sulfate	0.0
KCl	7	pH	8.6
$CaCl_2$	7	Phosphorus	0.05
$MgSO_4$	5	Hardness	23.6
$Al_2(SO_4)_3.18H_2O$	25		
$NaHCO_3$	168		

Sanitary Analysis

BOD, 5 day 20°C	190 ppm
Total Nitrogen	25.3 ppm
Orthophosphate phosphorus	5.5 ppm
Organic phosphorus	1.0 ppm
Alkalinity, total	143. ppm
pH	8.0

the salinity of the synthetic substrate. The pertinent chemical and radiochemical characteristics of these isotopes are given in Table 2. Phosphorus and iodine were selected for study primarily because they are the two most widely used isotopes in research and therapy. Strontium is an important and particularly hazardous constituent of the fission-products. It should be clearly understood that these three isotopes are not necessarily typical or representative of radioactive materials in general and that the results reported are only applicable to the elements studied.

The factors that may be expected to determine the degree of uptake of ionic radioactive materials may be either those characteristic of the waste, or those related to the operation of the activated sludge process. The most important waste properties include the carrier concentration (i.e., the concentration of stable isotopes of the radioactive element), the concentration of ions chemically similar to the radioisotope in question, the concentration of materials that may form insoluble precipitates with the radioisotope and its carrier, and the nutritional qualities of the waste, such as BOD, nitrogen, and phosphorus. An additional consideration unique to biological treatment is the biochemical role of the radioisotope and its stable counterpart. The operational parameters of the activated sludge process include the concentration of aeration solids, the period of aeration, and the organic loading per unit of time, expressed either as a ratio of the influent BOD per unit of aeration volume or as the BOD added per unit of volatile aeration solids present in the system.

The concentration of the radioisotope, expressed in microcuries (uc) or millicuries (mc) per milliliter, does not significantly influence the efficiency of the treatment process. This statement originates from two considerations. First: The level of radiation likely to be encountered in the application of biological waste treatment is below that necessary to damage or appreciably alter the functioning of the process. Second: The concentration of the radioisotope present in a waste will, in all probability, be chemically and biochemically insignificant and therefore can have little influence on the operation of the process. For example, one mc of phosphorus 32 per liter is equivalent to 3.5×10^{-6} ppm of phosphorus, a concentration negligible in comparison to that of the stable phosphorus found in sewage. It should be understood, however, that radioactive wastes may contain highly toxic nonradioactive materials that might preclude the utilization of biological treatment.

PROCEDURE

Radiological Aspects of Research

The level of radioactivity employed ranged from 5×10^{-3} to 20×10^{-3} uc per milliliter, depending on the expected removal efficiency of the system under study. The concentration of radioisotope was measured in counts per minute per ml and the uptake expressed as a percentage of the initial concentration. All counting was done with an end window Geiger-Muller tube. Care was taken to maintain constant geometry, and to correct for coincidence losses and radioactive decay in order that effluent counting data be comparable to assays of the untreated substrate. Self-absorption losses were evaluated and found to be insufficient to appreciably reduce the counting rate of most samples. In nearly all instances, it was possible to employ minimum counting rates of at least 60 counts per minute and to maintain the counting

Table 2. Properties of Radioisotopes Studied

Isotope	Half-Life Days	Characteristic Radiations	Chemical Form As Rec'd From Oak Ridge Nat'l Lab.	Recommended[5] Max. Permissible Concentration in Water uc/ml
P^{32}	14.3	β^- 1.72 Mev.	PO_4 in dilute HCl	2×10^{-4}
Sr^{89}	54.	β^- 1.5 Mev.	Sr^{++} as $SrCl_2$ in 0.5 N HCl	7×10^{-5}
I^{131}	8.0	β^- 0.60,0.25 Mev. γ 0.36, 0.64, 0.28 Mev.	I^- as NaI in basic Na_2SO_3 solution	3×10^{-5}

5. Subcommittee on Internal Dose of the National Committee on Radiation Protection, National Bureau of Standards Handbook 52. Maximum Permissible Amounts of Radioisotopes in Human Body and Maximum Permissible Concentrations in Air and Water. Superintendent of Documents, Washington 25, D.C. (April, 1953).

coefficient of variation to within 2 per cent. In determining the activity of sludge solids, however, the coefficient of variation of the mean of five observations often reached 15 per cent. This was probably caused by the lack of homogeneity of the original sample of which only small portions were counted to avoid excessively high counting rates.

Development of Activated Sludges

Activated sludge systems were developed at room temperature on the synthetic substrates from a typical domestic activated sludge. In order to study the effect of substrate constituents, sludges were grown on the standard substrate as shown in Table 1, on the standard substrate containing various phosphorus concentrations, and on a modified substrate having no nutrient broth but containing sufficient supplementary starch and urea to maintain a BOD of 190 ppm and a total nitrogen content of 25.3 ppm. The standard and modified substrates permitted the development of essentially identical activated sludges. Three to four weeks were required to achieve conditions of equilibrium as measured by the constancy of the sludge and effluent characteristics. Values for the Sludge Volume Index were generally below 200, although in several instances daily chlorination was necessary to prevent bulking.

Quantities of activated sludge were grown in 2.5 liter aeration tubes containing 1500 ml of mixed liquor. These units were fed twice daily by settling for 30 minutes, removing one liter of supernatant effluent, and adding one liter of the desired substrate. This procedure was equivalent to providing a 50 percent sludge return and resulted in a daily BOD loading of 15.8 pounds per 1000 cubic feet of aeration capacity. The aeration solids were maintained at a constant concentration by measuring the suspended solids each morning and wasting a calculated quantity of mixed liquor prior to the evening feeding. The uptake of radioisotopes in the batch scale studies was measured by using 155 ml quantities of mixed liquor. Fifty ml aliquots of sludge, developed in the 1.5 liter aeration tubes, were transferred to 300 ml Kjeldahl flasks. One hundred ml of substrate were added, followed by the addition of 5 ml of a solution containing a known concentration of the radioisotope under study. After six hours of aeration, the radioisotope remaining in the liquid phase was assayed and the percent uptake calculated. In the phosphorus 32 studies, the separation of the liquid effluent from the sludge solids was made by filtration, using Whatman filter paper No. 40. In subsequent work with strontium 89 and iodine 131, centrifuging was employed.

Pilot scale studies, in which the radioisotope was added continuously with the influent substrate, were carried out with the continous flow activated sludge units shown in Fig. 1. The 10 liter aeration tank provided a detention period of 8.33 hours at an influent rate of 20 ml per minute. The sludge was concentrated in the conical bottom of the settling basin and returned to the aeration tank at the rate of 10 ml per minute. The aeration solids were maintained nearly constant by wasting the return sludge for periods of from one to two hours daily, depending on the concentration of solids desired.

EXPERIMENTAL RESULTS

Phosphorus 32 Studies

The phosphorus requirements of the activated sludge process may be determined from the rate of development of new sludge solids and the phosphorus content of those solids. Treatment plants operating with high organic

loadings per unit of aeration solids produce large amounts of waste sludge and require that the influent waste contain considerable quantities of phosphorus in order to provide for the proper nutrition of the activated sludge. Helmers[2] has reported phosphorus requirements for industrial waste stabilization of 1.0 lb. per 100 lb. of BOD removed with the aeration solids at 1,500 ppm. Under a particular set of loading conditions, a given amount of organic sludge material should be expected to extract a nearly fixed quantity of phosphorus from the substrate, irrespective of the initial phosphorus concentration. In order to illustrate the influence of this relationship on the removal of phosphorus 32, a batch study was carried out with various initial phosphorus concentrations. The result of this experiment is shown in Figure 2. The lower portion of the curve is hyperbolic as would be expected if a fixed quantity of phosphorus was taken up by the sludge solids.

The effect of organic loading on the phosphorus requirements is illustrated in Figure 3. In order to maintain a definite initial phosphorus concentration, the modified substrate was employed. Various strengths, ranging up to eight times the normal, were used. After the usual six hour period of aeration, samples of the effluent were analyzed for stable soluble phosphorus and phosphorus 32. Although the stable and radioactive phosphorus were added in an identical chemical form,i.e., as the orthophosphate, the removal of phosphorus 32 exceeded that of the stable phosphate at all loadings. Ordinarily, under such circumstances, identical removals of the stable and radioactive forms would be expected. This seeming anomaly can only be explained by the existence of a dynamic equilibrium between the phosphorus in solution and the exchangeable organic phosphorus within the activated sludge. Phosphorus 32 was removed by incorporation into new protoplasm and by exchange with stable phosphorus present in existing cell substance. As the BOD loading was increased, growth also increased and incorporation in this new growth became the predominating mechanism of uptake. At zero BOD loading, although no stable phosphorus was required, 15 percent of the phosphorus 32 was removed by exchange. In a similar batch study employing a sludge developed on the standard substrate containing 1.5 ppm stable phosphorus, phosphorus 32 removals in excess of 98 percent were obtained at all loadings above 20 pounds of BOD per day per 1000 cu.ft. of aeration capacity.

The influence of aeration solids on phosphorus 32 uptake, as measured by the batch study technique, is illustrated in Figure 4. These data also require interpretation in the light of the isotopic exchange mechanism of assimilation. The BOD loading was maintained constant at 31.6 pounds per day per 1000 cu. ft. of aeration capacity. At low aeration solids concentrations the relatively high rate of growth is responsible for a considerable portion of the radioactive phosphorus removal. At 2400 ppm aeration solids, very little growth took place, indicating that the exchange phenomenon represented the principal mechanism.

The relationship of aeration period to phosphorus 32 uptake at various combinations of aeration solids and stable phosphorus concentrations is illustrated in Figure 5. These studies were made on a batch scale which permitted the exchange assimilation of phosphorus 32. For this reason, the higher suspended

2. "Nutritional Requirements in the Biological Stabilization of Industrial Wastes," by E. N. Helmers, J. D. Frame, A. E. Greenberg, and C. N. Sawyer, Sewage and Industrial Wastes, Vol. 23, No. 7, July, 1951.

solids provided slightly greater phosphorus 32 removals at all aeration periods. It may be observed that the exchange reaction was essentially complete within a 10-hour period and that phosphorus 32 may actually be released into solution if aeration is prolonged. When the stable phosphorus concentration became limiting from the nutritional standpoint, the rate of phosphorus 32 uptake was very rapid. At higher aeration solids concentrations the rate was further increased by the greater quantity of stable phosphorus interchanging between the liquid and solid phases of the system.

To appreciate the significance of the data presented in the preceding paragraphs, two types of radioactive waste problems must be considered. In the case of a treatment plant receiving occasional transient concentrations of phosphorus 32, maximum removals could be achieved by maintaining a high aeration solids level and, if operating conditions permit, a minimal carrier phosphorus concentration. The exchangeable reservoir of stable phosphorus within the activated sludge solids would serve to reduce peak phosphorus 32 levels and to release the assimilated radioisotope into the effluent gradually. The function of a plant operated in this manner would be to provide temporary storage, thus permitting greater dilution in the receiving body of water. The industrial treatment plant receiving phosphorus 32 continuously should be operated so as to achieve a maximum rate of sludge growth, i.e. by employing the maximum practical BOD loading per unit of aeration solids. Under such operating conditions the phosphorus requirement will be at a maximum, but only sufficient stable phosphorus should be provided to meet the minimum nutritional needs of the activated sludge.

Continous Flow Pilot Plant Studies

When the stable and radioactive phosphorus are present in identical chemical forms, a so-called "tracer" relationship exists with the phosphorus 32 tracing the movement of the stable or carrier form of the isotope. The ratio of the phosphorus 32 concentration to that of the stable phosphorus, expressed in any convenient units, is termed the specific activity. The tracer principle may be applied to the problem of predicting the phosphorus 32 uptake in a continous flow activated sludge plant by determining the stable phosphorus removal chemically. If it may be assumed that the major portion of the influent phosphorus is in a form available for metabolism, then a tracer relationship is approximated and the percent reduction in stable phosphorus will equal the percent reduction in phosphorus 32. The calculated uptake will represent a minimum and be strictly correct only for a plant treating a waste containing a constant level of radioactivity. In cases of transient comtamination, isotopic exhange will result in greater reductions that may only be evaluated with some difficulty.

In order to demonstrate this tracer principle a continous flow pilot scale study was carried out over a three week period with the results shown in Figure 6. Using the modified substrate, the pilot unit was brought to a steady state by continuous operation for a four-week period prior to adding the radioactive phosphorus to the influent. The phosphorus 32 level in the effluent became nearly constant, ranging between 70 and 80 percent of the initial, after approximatley eight days of operation. The specific activities of the effluent and sludge solids were determined periodically and have been plotted against time on Figure 6. After 10 days of adding phosphorus 32 the influent the specific activities of the influent, effluent, and sludge solids became essentially equal. The percent removal of phosphorus 32 then equaled that of the stable phosphorus and a complete tracer relationship existed. A similar study was

made using the standard substrate. The results were essentially identical, indicating that the organic phosphorus present in the nutrient broth was biologically available to the activated sludge organisms.

Strontium 89 Studies

The parameters investigated with respect to strontium 89 uptake were the aeration period, the aeration solids concentration, the BOD loading and the concentration of stable strontium, calcium, magnesium, aluminum, and orthophosphate phosphorus. All experiments were performed as batch studies employing the modified substrate for the development of the activated sludges, but using distilled water in preparing the modified substrate for the actual tests using strontium 89. Under these conditions the influence of variations of the mineral content of the tap water was minimized, and a closer control of the parameters under study was possible. The initial concentrations of the more important ions present in the 155 ml uuantities of mixed liquor used are given below:

Element	Sr	Ca	Mg	Al	P
Concentration-ppm.	0.3	4	1	0.3	5.9

A comparison of Figures 5 and 7 indicates that the uptake of strontium 89 is considerably more rapid than that of phosphorus 32. This marked difference in the time rates of removal strongly suggests that a surface adsorption mechanism is operative in strontium 89 uptake, since the diffusional processes necessarily involved in assimilative incorporation require much greater periods of time. It is an established fact that surface adsorption phenomena generally reach near completion within a fifteen minute reaction period. Higher BOD loadings gave the same strontium 89 uptake after 15-minute and 6-hour aeration periods (Figure 9), in spite of the fact that the removal of high concentrations of BOD requires considerably more than 15 aeration.

The effect of the aeration solids level was studied at calcium concentrations of 4 and 47 ppm, respectively. At a calcium concentration of 47 ppm, Figure 8 shows a nearly linear relationship between strontium 89 uptake and suspended solids concentration. At the lower calcium level, strontium 89 uptake is considerably greater and the removal-solids relationship is no longer linear. Strontium and calcium ions are adjacent menbers of the lyotropic series and are believed to behave in biologically similar fashions. If it may be assumed that calcium and strontium freely undergo isotopic exchange, the influence of suspended solids and calcium on strontium 89 uptake is readily explained in terms of an exchange adsorption mechanism. When the ionic calcium is low, the greater portion of the calcium in the system is present in the solid phase and a higher degree of strontium 89 uptake occurs. As the aeration solids are increased the removal may approach, but of course never reaches, one hundred percent.

The influence of BOD loading on uptake is illustrated in Figure 9. In contradistinction to the effect of the loading parameter on phosphorus 32 removal, the effect in the case of strontium, if actually significant, was to bring about a reduction in uptake. The relationship of growth to BOD loading is also shown in Figure 9. The uptake of strontium did not increase, in spite of the fact that a 50 percent increased in the aeration solids took place. It is difficult to account for these results since the newly developed protoplasm may be expected to require additional calcium and presumably strontium.

The influence of carrier strontium, as illustrated in Figure 10, is similar

to that of calcium and magnesium as shown in Figure 11. It is apparent from these data, and from the aeration time study, that the three ions are most probably taken up by surface adsorption in approximately similar quantities and that a greater amount of any one may be expected to reduce the removal of strontium 89. The manner in which the aluminum ion influences the uptake of strontium 89 is of particular interest. At low aluminum concentrations, the formation of a hydrous aluminum oxide probably accounts for the absence of interference with strontium 89 uptake. As the aluminum concentration is increased, the trivalent aluminum ion exerts a marked influence, eliminating practically all removal of strontium by the sludge solids. This relationship may be taken as further evidence of an exchange adsorption mechanism of strontium 89 uptake.

The possibility of strontium 89 forming a precipitate with the orthophosphate phosphorus or being precipitated with calcium or magnesium phosphate was investigated with the modified substrate and a range of phosphorus concentrations from 1.6 to 10.7 ppm. The results of a series of these tests, in which the initial calcium concentration was 8.0 ppm, are shown below:

Initial Soluble Phosphorus, ppm	1.57	2.33	4.50	7.07	8.10	10.67
Strontium 89 Uptake, percent	51	52	51	52	54	50

No significant change resulted from increasing the phosphorus concentration within the above limits. It should be noted, however, that calcium and strontium phosphates are quite insoluble and, at high pH, may be used to remove strontium 89 by carrier precipitation.

Iodine 131 Studies

In planning the investigation on the removal of radioactive iodine, very little information was available pertaining to the role of stable iodine in the metabolism of microorganisms. The iodine present in surface waters is generally less than 0.010 ppm. It is probable that very little is necessary for the development of activated sludges and that its function may be that of a trace element. Straub[3], quoting Carter[3a], has reported iodine 131 removals of 85 percent by trickling filters and secondary sedimentation, while Belcher[4] found uptakes of 5 percent by trickling filters and 11 to 14 percent by the activated sludge process. The variations in these data indicate that, although iodine may bot be required in large quantities by microorganisms, the conditions of biological treatment may exert a considerable influence on the degree of uptake.

The parameters investigated in the batch studies included aeration solids, BOD loading, aeration period, and the carrier iodine and chloride ion concentrations. The activated sludges employed were developed on the modified substrate. In making up the feed solutions for the actual batch studies, the

3. "Effect of Radioactive Materials on Environmental Health", by C. P. Straub, Public Health Reports, Vol. 67, No. 3, Page 298, March, 1952.

3a. "Study on the Removal of Radioactive Iodine (I^{131}) by Laboratory Sewage Trickling Filters." Masters Thesis, Georgia Institute of Technology, Atlanta, 1951. See also: Sewage and Industrial Wastes, Vol. 25, No. 5, May 1953.

4. "Experimental Studies on the Fate of Radioactive Materials in Sewage Treatment", by E. H. Belcher, Annual Conference, Institue of Sewage Purification, 1951 (British).

modified substrate was prepared with distilled water in order to minimize the variation in iodine and chloride concentrations that might result from changes in the mineral constituents of the tap water. The iodide concentration present in the mixed liquor was in all instances estimated to be less than 0.001 ppm, and the chloride concentration, except where noted to the contrary, was 30 ppm.

The effects of the aeration period and the aeration solids concentration on iodine 131 uptake are shown in Figure 12. In order to evaluate any removal of radioiodine by adsorption on glassware or by chemical precipitation, a parallel time study was made with the modified substrate but in the absence of activated sludge. No uptake was observed throughout a 24-hour period, ruling out this possibility within the system under study. From 2 to 4 hours were required to bring about maximum removals, a period somewhat shorter than that for phosphorus 32, but considerably longer than for strontium 89. This time-uptake relationship is indicative of an absorptive or diffusional assimilation mechanism rather than simple surface exchange adsorption. In this respect, iodine is similar to orthophosphate phosphorus. After four hours of aeration, the aeration solids concentrations studied had very little influence on the iodine 131 uptake. The increases in aeration solids at 590 ppm and 1430 ppm were approximately 20 and 6 percent, respectively. These data indicate a reduction, at high aeration solids, in the exchange capacity of the activated sludge per unit of sludge solids.

The influence of BOD loading is illustrated in Figure 9. Iodine 131 is very similar to strontium 89 from the standpoint of the organic loading parameter in that high loadings brought about slightly reduced uptakes. The aeration solids concentration was 1570 ppm at the beginning of the aeration period and, at high organic loadings, increased to nearly 2400 ppm during the six hour aeration period. Here we have the same anomalous relationship as observed in the strontium studies; namely, that the newly formed aeration solids do not have the capacity to remove iodine from the substrate.

The carrier iodide concentration is shown in Figure 13 to be the most critical parameter in the iodine 131 uptake scheme. This, in all probability, accounts for the variance reported in the literature. Within the range of iodide concentrations generally found in natural surface waters, the radioiodine removal varied from 15 to 98 percent. Increasing the chloride concentration from 30 to 60 ppm only reduced the iodine 131 uptake from 98.6 to 95.0 percent in spite of the fact that the stable iodide concentration was probably less than 0.001 ppm. If the mechanism of iodine removal were one of physical adsorption, this would not be the case. Carrier iodide concentrations less than 0.01 ppm had little influence on the iodine 131 uptake, while concentrations from 0.5 to 100 ppm resulted in essentially zero uptake.

The removal of phosphorus 32 is directly related to the BOD loading, increasing at high loadings per unit of aeration solids. The relationship of organic loading to strontium 89 and iodine 131 removal is not entirely clear, although the batch studies reported herein demonstrated little or no significant correlation between loading and isotope uptake.

DISCUSSION AND CONCLUSIONS

The application of the activated sludge process to the concentration of the radioisotopes of phosphorus, iodine, and strontium from liquid wastes has been investigated. The elements studied were employed as soluble inorganic compounds and were added to a synthetic sewage to provide a simple radio-

active waste of known composition. The chemical concentrations of the radioisotopes were sufficiently low that they may not be expected to exert any significant influence on the operation of the treatment process. For this reason, the results reported herein are applicable to any combination of the three radioisotopes studied, providing that they are present at approximately the same concentrations as that used in this study and in a waste of composition similar to that employed. If the radioisotopes are incorporated in insoluble inorganic compounds, they cannot be expected to participate in biochemical reactions; however, granular and colloidal radioactive agglomerates are more easily separated from the liquid phase than the ionic forms.

In all instances the most important parameter was the concentration of the stable or carrier form of the isotope under study. When high concentrations of the carrier were present, only minor removals of the radioactive elements were obtained with the activated sludge process. In the strontium 89 investigation, increasing concentrations of magnesium, calcium and aluminum also reduced the uptake of radioactivity. It may be concluded that biological treatment methods have limited application to the concentration of mono- and di-valent ionic radioelements from highly mineral wastes. In such cases, chemical precipitation has a greater likelihood of being successfull. Tri-and tetra-valent ionic radioisotopes are probably more highly adsorbed and are likely to form hydrous oxides under the conditions suitable for the development of biological sludges. In a radioactive organic waste of high salinity, it would probably be desirable to provide series treatment with both biological and chemical treatment units.

The removal of phosphorus 32 is directly related to the carrier phosphorus requirements of the biological system. These requirements are greatest under conditions of BOD loading resulting in a maximum rate of sludge growth. The relationship of BOD loading to strontium 89 and iodine 131 removal is not entirely clear since increased loadings did not result in greater radioisotope uptakes.

The removal of radioactive phosphorus from domestic sewage by the activated sludge process may be rather closely estimated if the carrier phosphorus content of the waste and the organic loading of the aeration tank are known. This conclusion is in accord with chemical tracer theory and has been borne out experimentally by continuous pilot scale studies. The existence of a dynamic equilibrium between the soluble and organic phosphorus present in the aeration solids makes available for isotopic exchange the large quantity of stable phosphorus present in the sludge solids. This phenomenon serves to reduce the level of radiophosphorus in the effluent of a municipal activated sludge plant receiving occasional transient concentrations of phosphorus 32. Conversely, it also brings about the gradual release of radiophosphorus from the sludge, thereby reducing the possible hazards from subsequent utilization of the waste sludge.

ACKNOWLEDGEMENTS

This investigation is a part of the waste research program sponsored by the United States Atomic Energy Commission under the guidance of Mr. Arthur E. Gorman. The work was performed by the Sanitary Engineering Research Project of the University of California under the directorship of Professor H. B. Gotaas. The assistance of Dr. Jerome Thomas and Mr. A. E. Greenberg is gratefully acknowledged.

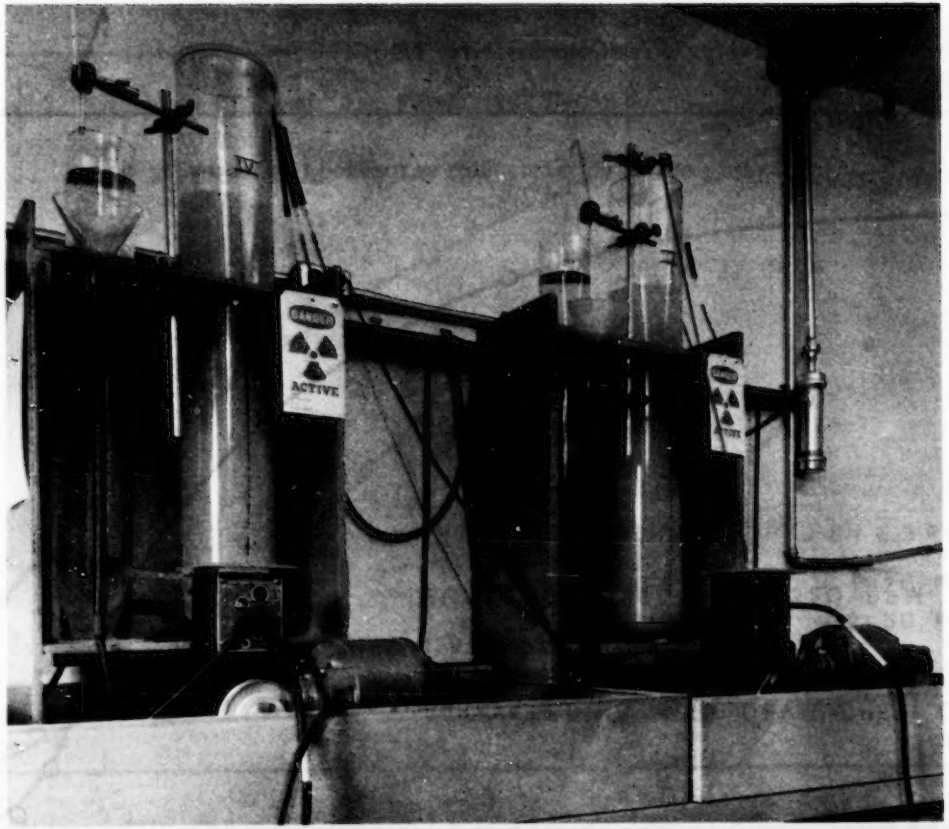


Fig. 1. Continuous Flow Pilot Scale Activated Sludge Units

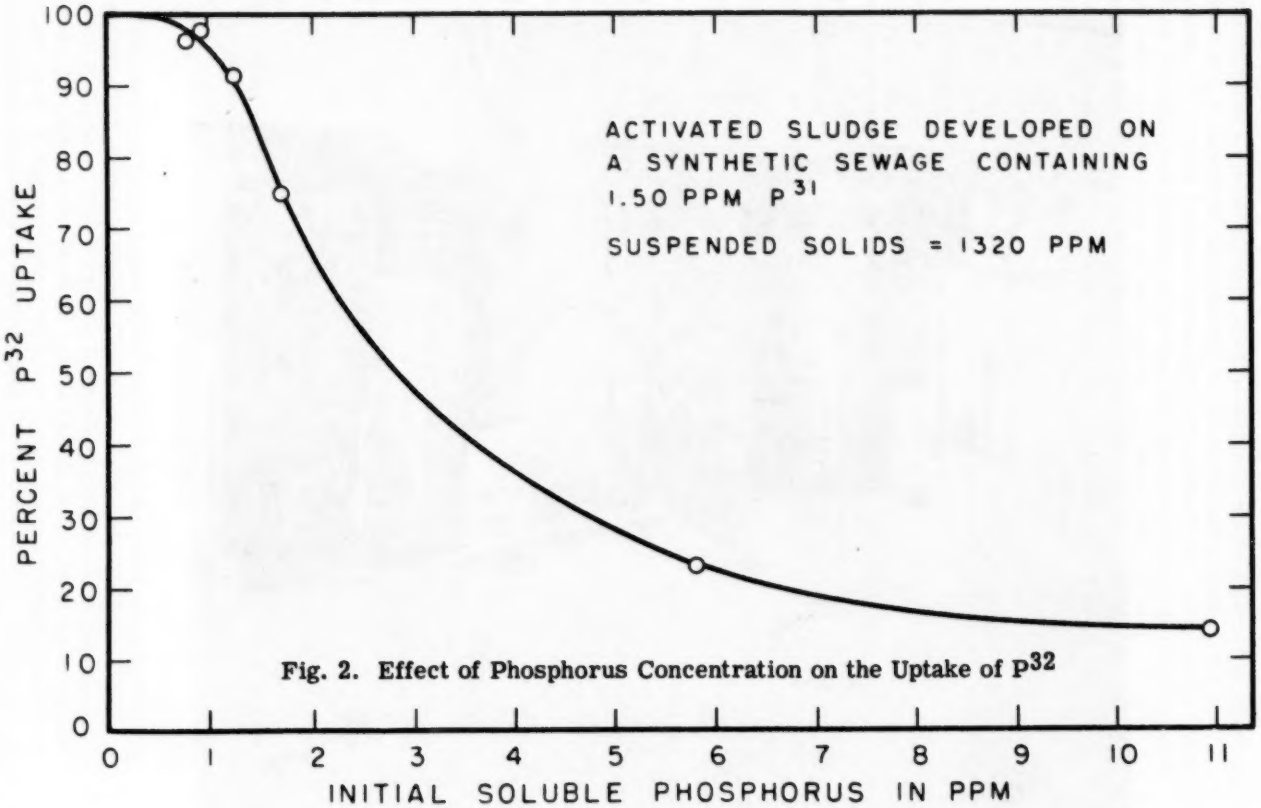


Fig. 2. Effect of Phosphorus Concentration on the Uptake of P^{32}

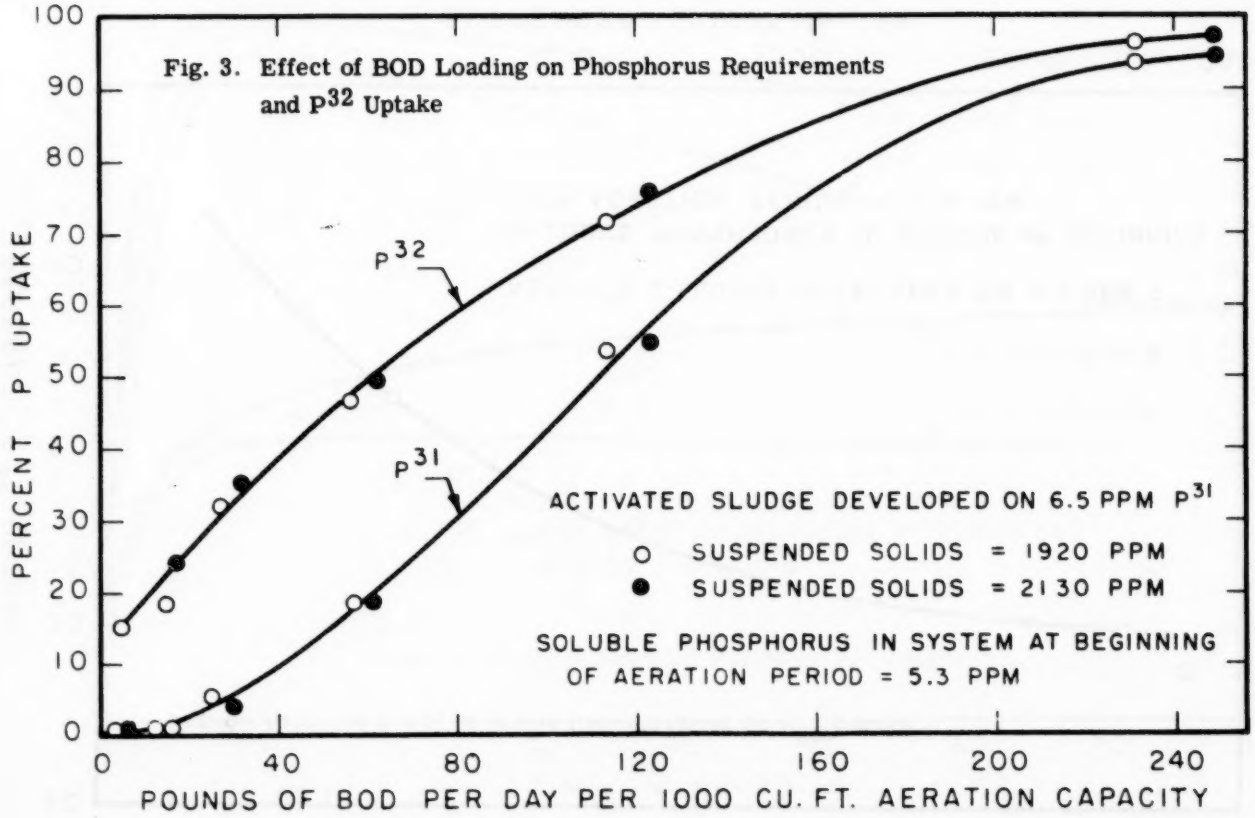
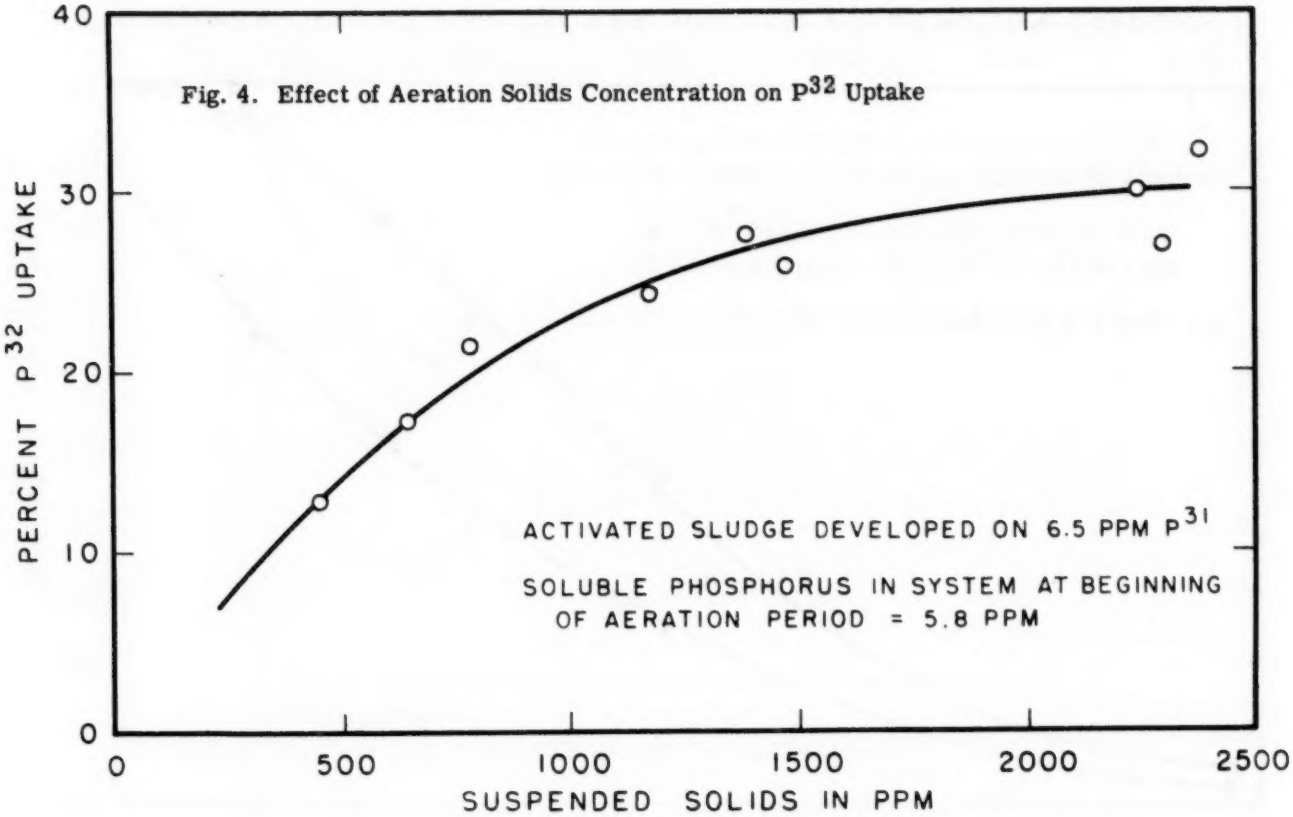


Fig. 3. Effect of BOD Loading on Phosphorus Requirements and P^{32} Uptake

Fig. 4. Effect of Aeration Solids Concentration on P^{32} Uptake



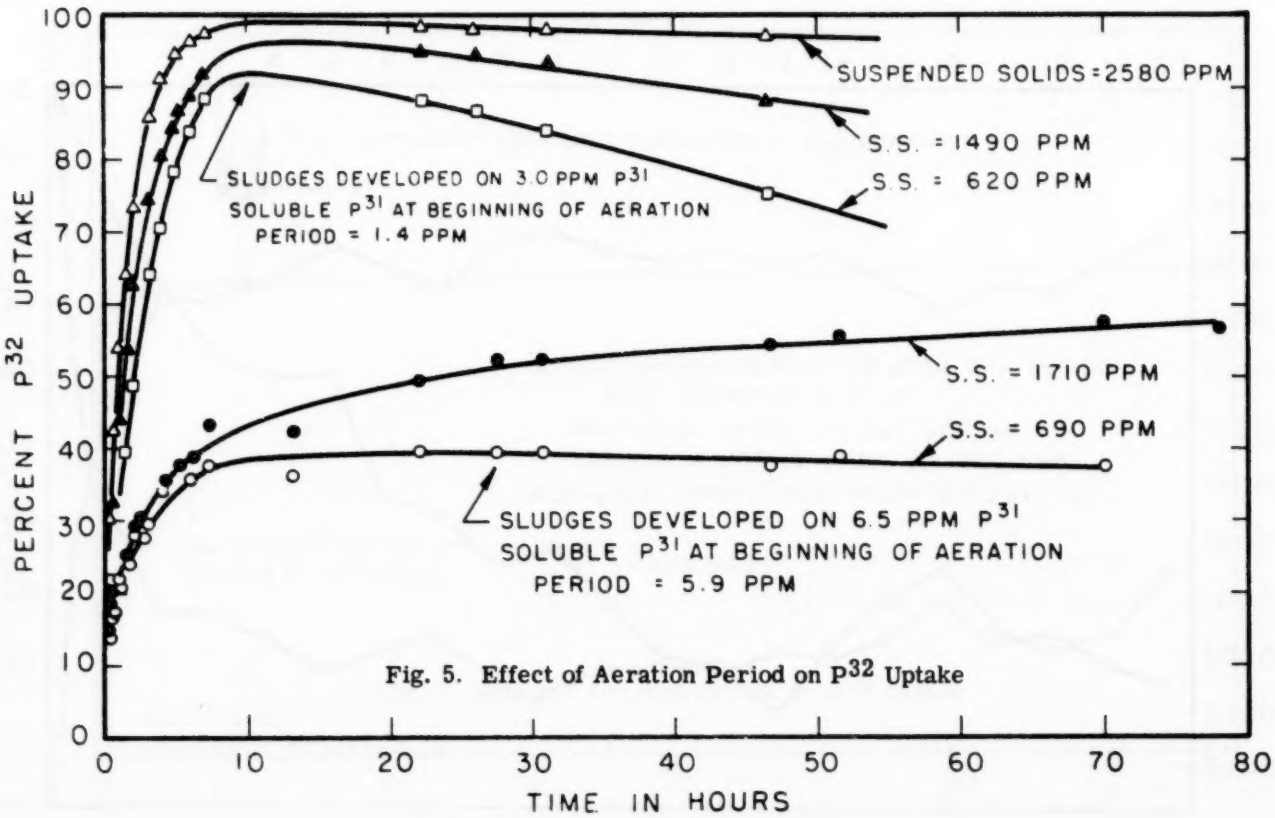

PERCENT P32 UPTAKE
TIME IN HOURS
SUSPENDED SOLIDS = 2580 PPM
S.S. = 1490 PPM
S.S. = 620 PPM
SLUDGES DEVELOPED ON 3.0 PPM P31
SOLUBLE P31 AT BEGINNING OF AERATION
PERIOD = 1.4 PPM
S.S. = 1710 PPM
S.S. = 690 PPM
SLUDGES DEVELOPED ON 6.5 PPM P31
SOLUBLE P31 AT BEGINNING OF AERATION
PERIOD = 5.9 PPM

Fig. 5. Effect of Aeration Period on P^{32} Uptake

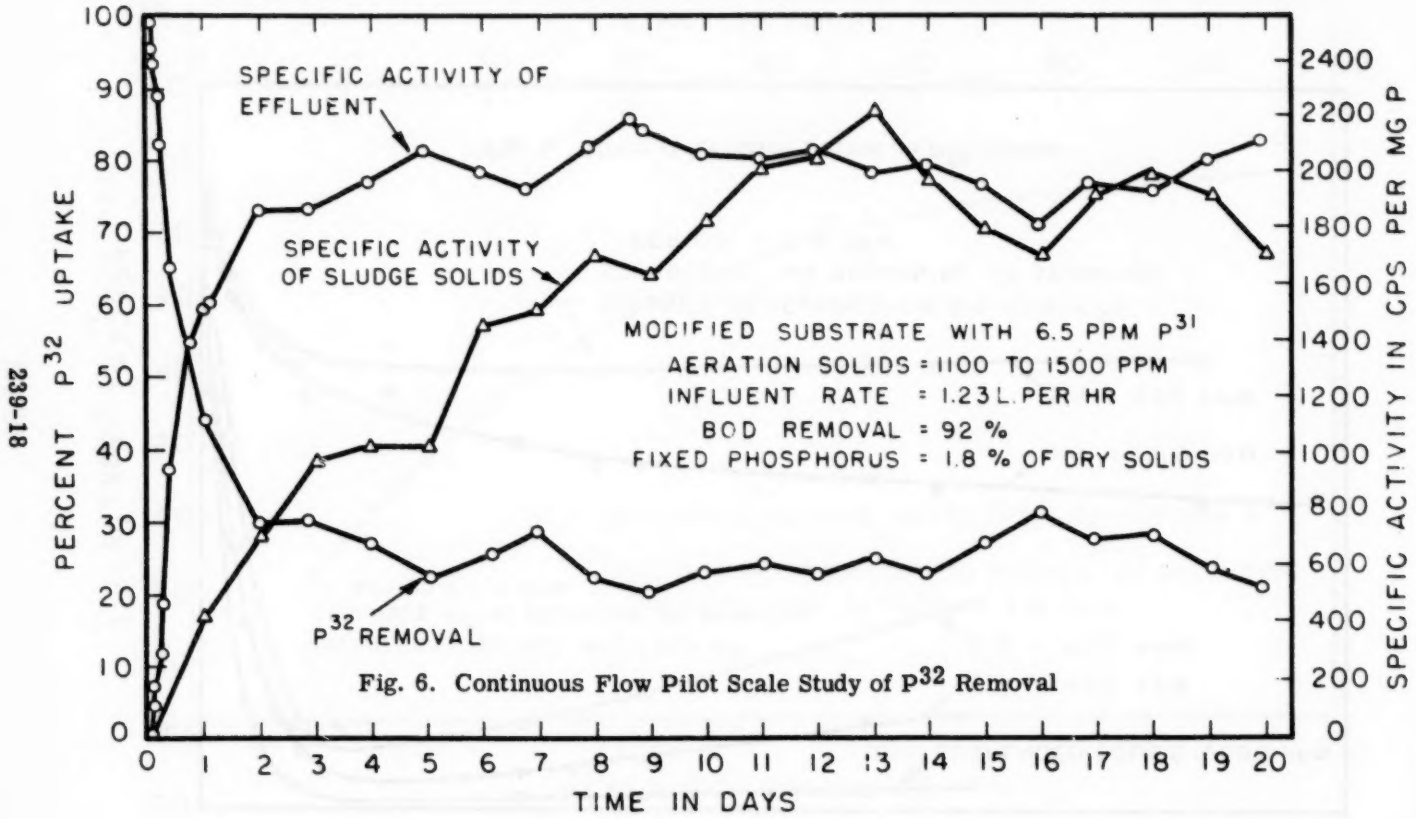

SPECIFIC ACTIVITY OF EFFLUENT
SPECIFIC ACTIVITY OF SLUDGE SOLIDS
MODIFIED SUBSTRATE WITH 6.5 PPM P31
AERATION SOLIDS = 1100 TO 1500 PPM
INFLUENT RATE = 1.23 L. PER HR
BOD REMOVAL = 92 %
FIXED PHOSPHORUS = 1.8 % OF DRY SOLIDS
P32 REMOVAL
PERCENT P32 UPTAKE
SPECIFIC ACTIVITY IN CPS PER MG P
TIME IN DAYS
0 10 20 30 40 50 60 70 80 90 100
0 200 400 600 800 1000 1200 1400 1600 1800 2000 2200 2400
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20

Fig. 6. Continuous Flow Pilot Scale Study of P^{32} Removal

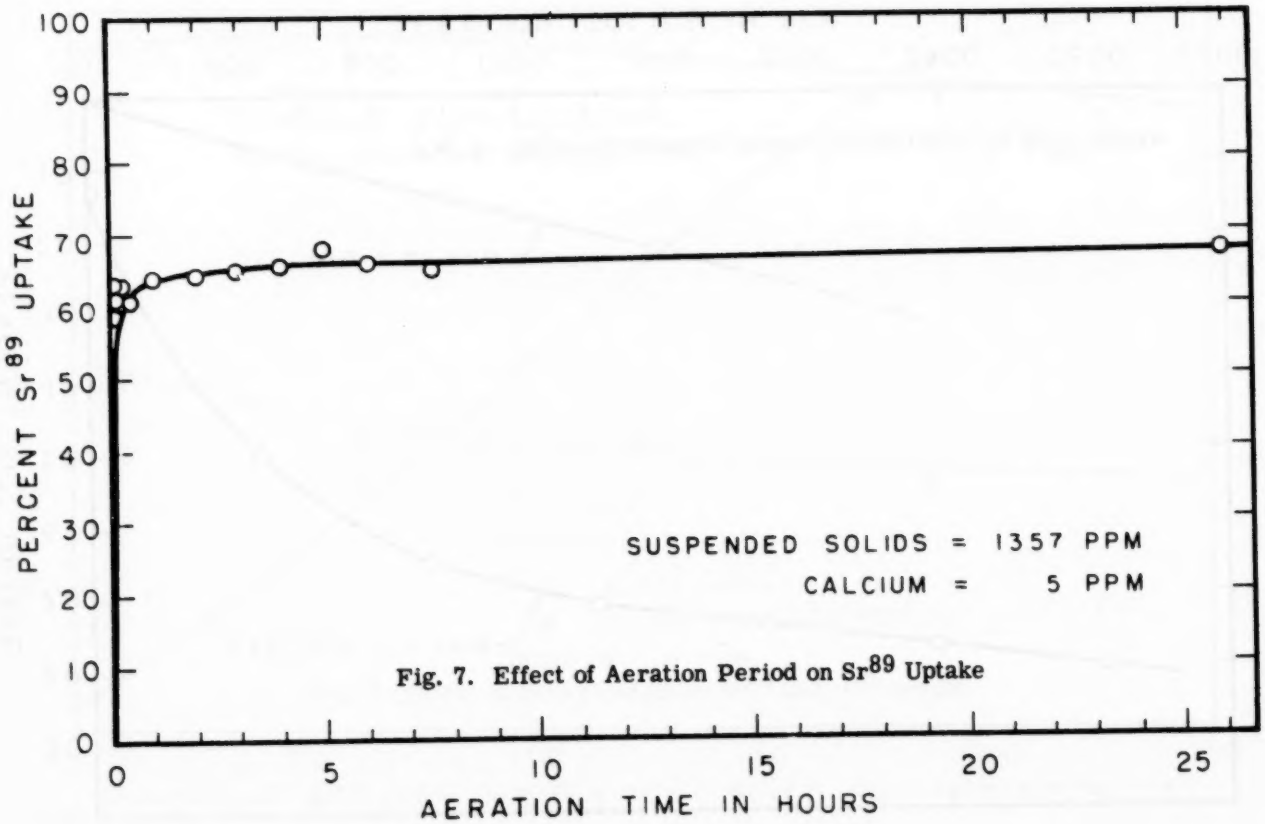


Fig. 7. Effect of Aeration Period on Sr^{89} Uptake

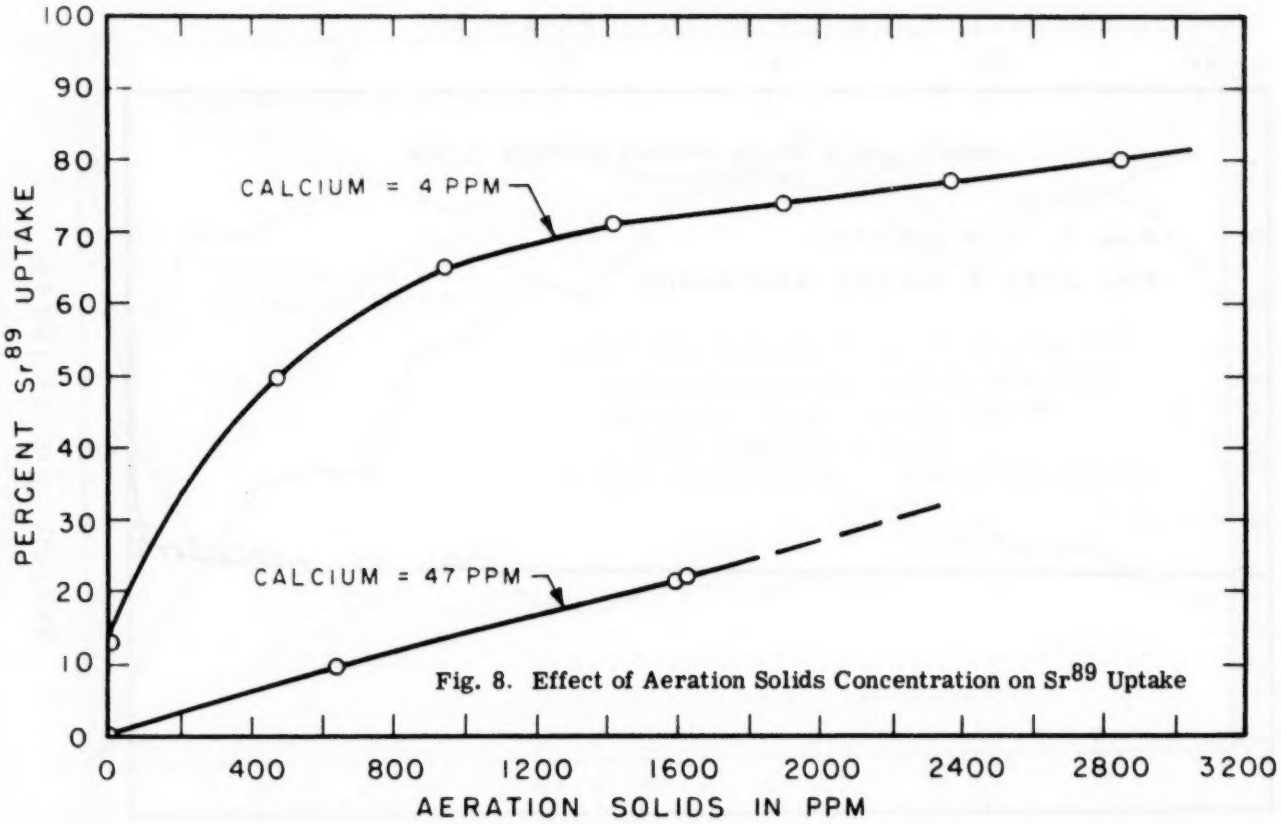


Fig. 8. Effect of Aeration Solids Concentration on Sr^{89} Uptake

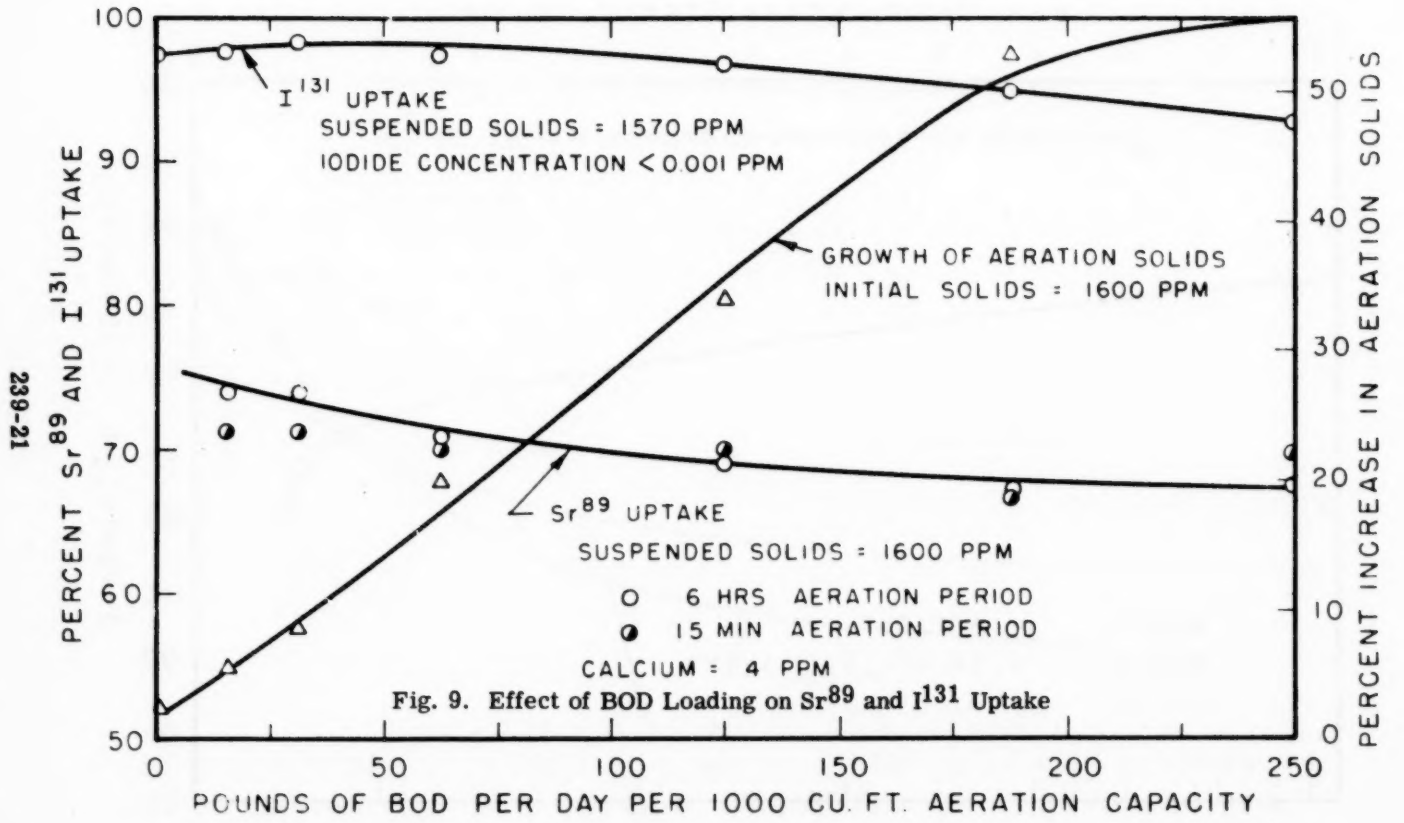


Fig. 9. Effect of BOD Loading on Sr^{89} and I^{131} Uptake

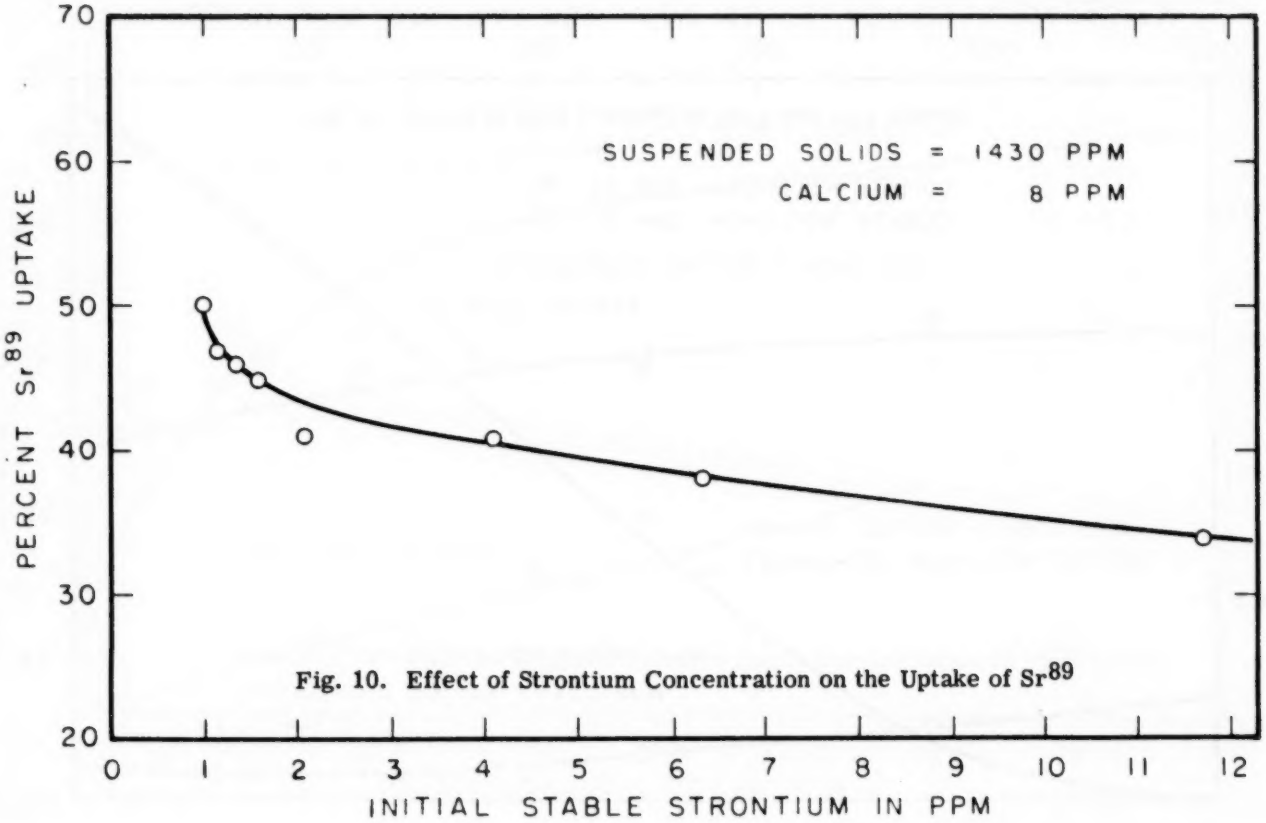
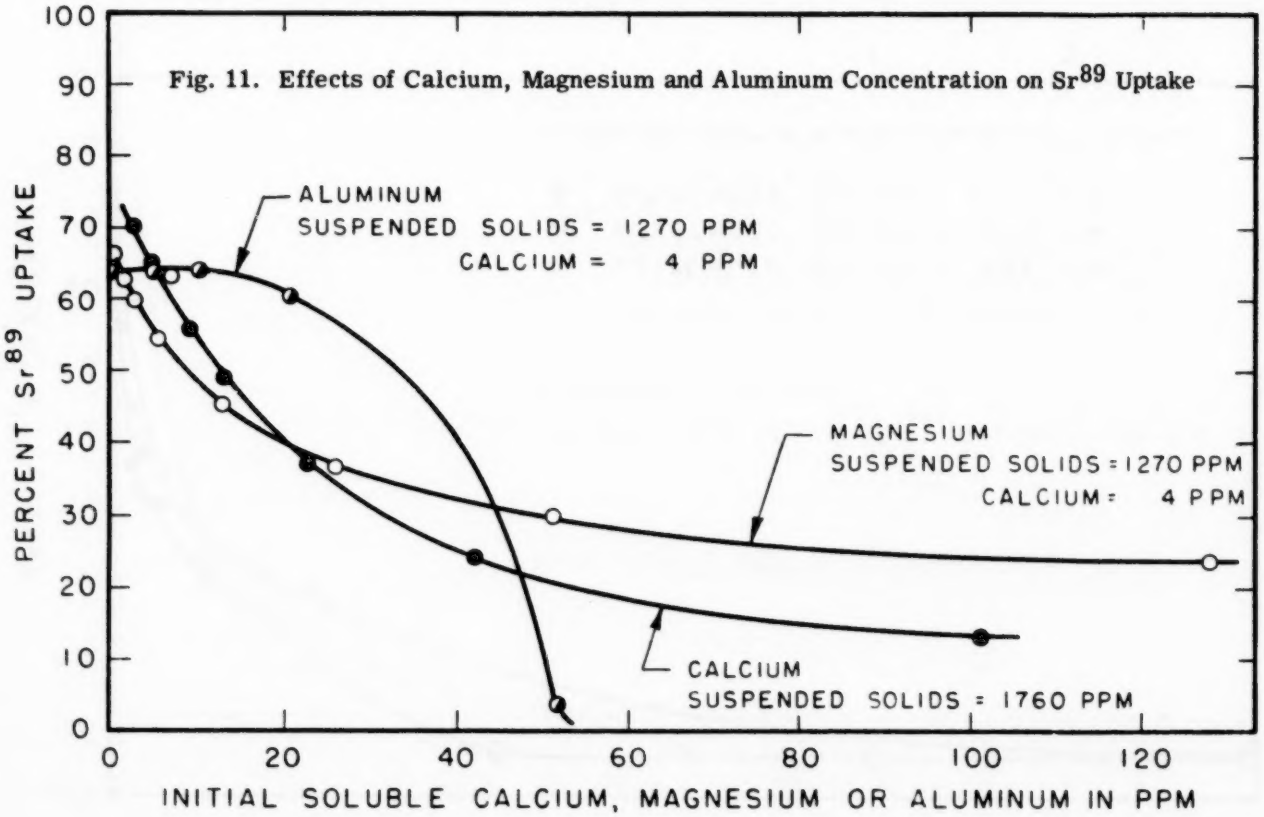


Fig. 10. Effect of Strontium Concentration on the Uptake of Sr^{89}

Fig. 11. Effects of Calcium, Magnesium and Aluminum Concentration on Sr^{89} Uptake



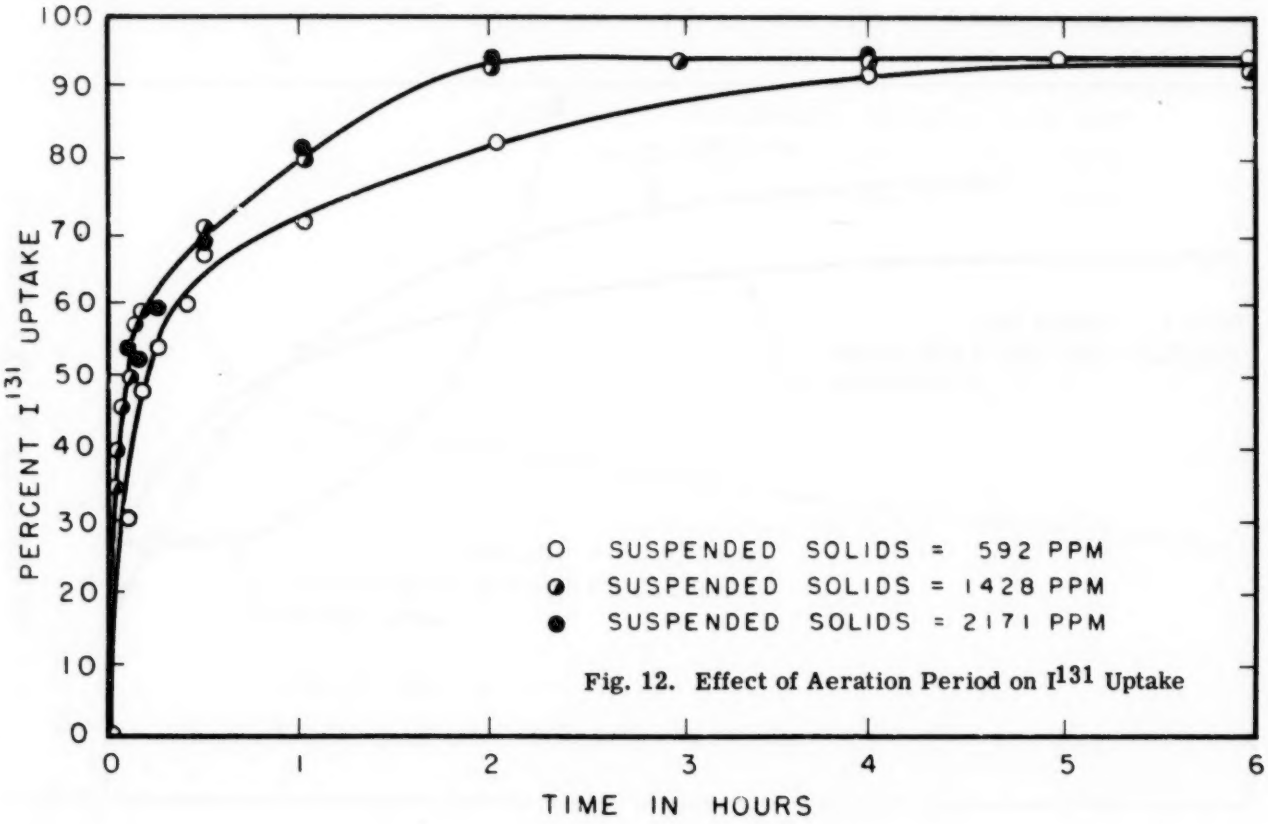


Fig. 12. Effect of Aeration Period on I^{131} Uptake

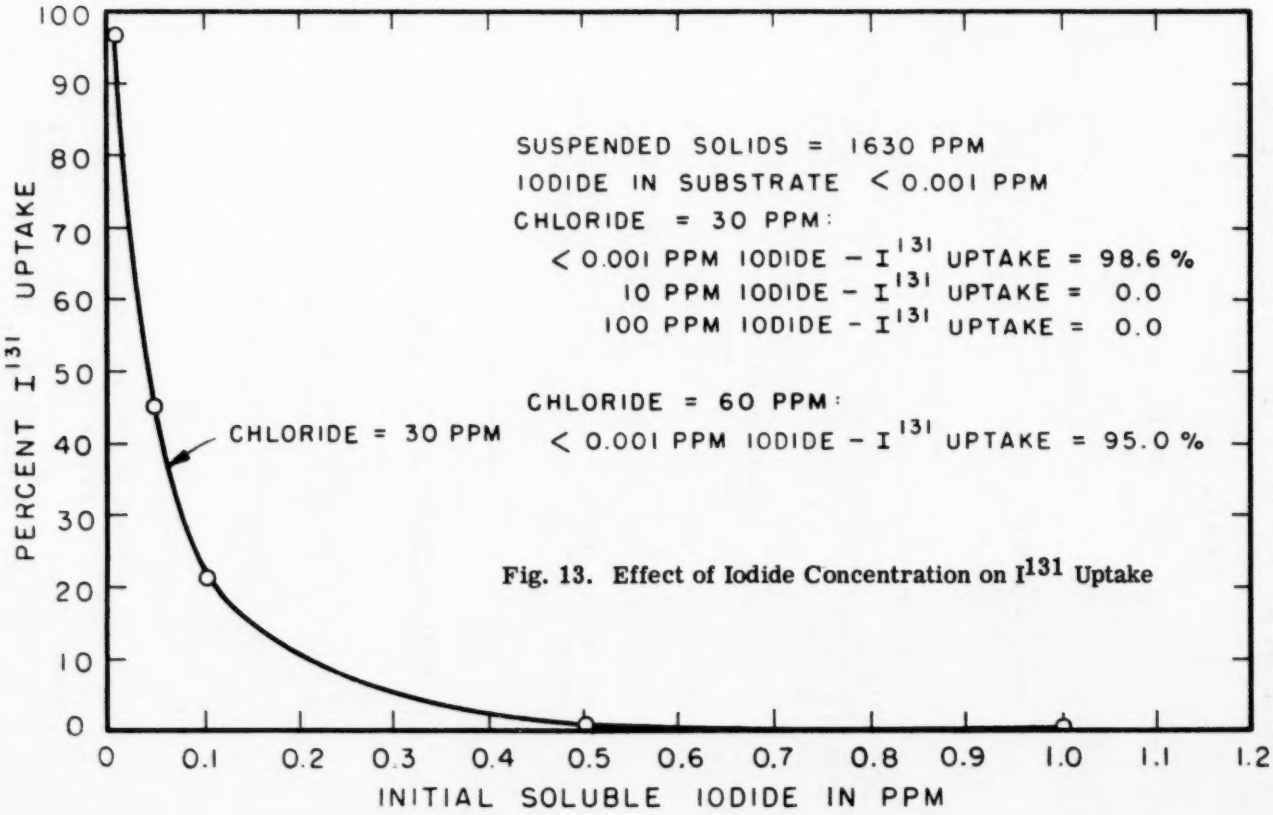

PERCENT I^{131} UPTAKE
INITIAL SOLUBLE IODIDE IN PPM
SUSPENDED SOLIDS = 1630 PPM
IODIDE IN SUBSTRATE < 0.001 PPM
CHLORIDE = 30 PPM:
< 0.001 PPM IODIDE - I^{131} UPTAKE = 98.6 %
10 PPM IODIDE - I^{131} UPTAKE = 0.0
100 PPM IODIDE - I^{131} UPTAKE = 0.0
CHLORIDE = 60 PPM:
< 0.001 PPM IODIDE - I^{131} UPTAKE = 95.0 %
CHLORIDE = 30 PPM

Fig. 13. Effect of Iodide Concentration on I^{131} Uptake

AMERICAN SOCIETY OF CIVIL ENGINEERS

OFFICERS FOR 1953

PRESIDENT

WALTER LEROY HUBER

VICE-PRESIDENTS

Term expires October, 1953:
GEORGE W. BURPEE
A. M. RAWN

Term expires October, 1954:
EDMUND FRIEDMAN
G. BROOKS EARNEST

DIRECTORS

Term expires October, 1953:
KIRBY SMITH
FRANCIS S. FRIEL
WALLACE L. CHADWICK
NORMAN R. MOORE
BURTON G. DWYRE
LOUIS R. HOWSON

Term expires October, 1954:
WALTER D. BINGER
FRANK A. MARSTON
GEORGE W. McALPIN
JAMES A. HIGGS
I. C. STEELE
WARREN W. PARKS

Term expires October, 1955:
CHARLES B. MOLINEAUX
MERCEL J. SHELTON
A. A. K. BOOTH
CARL G. PAULSEN
LLOYD D. KNAPP
GLENN W. HOLCOMB
FRANCIS M. DAWSON

PAST-PRESIDENTS

Members of the Board

GAIL A. HATHAWAY
CARLTON S. PROCTOR

TREASURER

CHARLES E. TROUT

EXECUTIVE SECRETARY

WILLIAM N. CAREY

ASSISTANT TREASURER

GEORGE W. BURPEE

ASSISTANT SECRETARY

E. L. CHANDLER

PROCEEDINGS OF THE SOCIETY

HAROLD T. LARSEN
Manager of Technical Publications

DEFOREST A. MATTESON, JR.
Assoc. Editor of Technical Publications

PAUL A. PARISI
Asst. Editor of Technical Publications

COMMITTEE ON PUBLICATIONS

LOUIS R. HOWSON

FRANCIS S. FRIEL
GLENN W. HOLCOMB
I. C. STEELE
FRANK A. MARSTON
NORMAN R. MOORE

* Readers are urged to submit discussion applying to current papers. Forty free Separates per year are allotted to members. Mail the coupon order form found in the current issue of *Civil Engineering*.